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Hisashi ISAKA et al. : **Docket No. 2002-0382A**  
Serial No. 10/088,265 : **Group Art Unit 1615**  
Filed March 18, 2002 : **Examiner P. Short**

POLYORTHOESTER AND CURABLE  
COMPOSITION CONTAINING THE SAME

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**DECLARATION**

Commissioner for Patents  
P. O. Box 1450  
Alexandria, VA 22313-1450

Sir:

I, Akiko KOJIMA, declare and say:

that I am thoroughly conversant in both the Japanese and English languages;

that I am presently engaged as a translator in these languages;

that the attached document represents a true English translation of the  
Japanese Priority Application No. 337837/99, filed November 29, 1999.

I further declare that all statements made herein of my own knowledge are true  
and that all statements made on information and belief are believed to be true; and  
further that these statements were made with the knowledge that willful false  
statements and the like so made are punishable by fine or imprisonment, or both,  
under Section 1001 of Title 18 of the United States Code, and that such willful false  
statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this 10th day of May, 2004.

*Akiko Kojima*

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Akiko KOJIMA



(TRANSLATION)

PATENT OFFICE  
JAPANESE GOVERNMENT

This is to certify that the annexed is a true copy of the following  
application as filed with this Office.

Date of Application : November 29, 1999

Application Number: Patent Application No. 337837/99

Applicant(s) : Kansai Paint Co., Ltd.

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Kozo OIKAWA

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Commissioner,  
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[Name of Object]	Specification	1
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[Name of Object]	Abstract	1
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[Proofing] Required

[Document Name] Specification

[Title of the Invention] Curable Composition

[Claims]

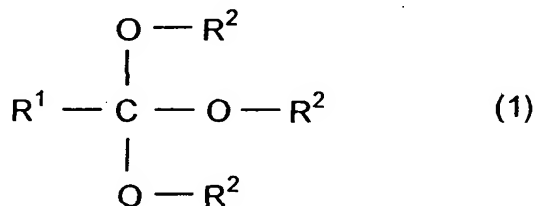
[Claim 1] A curable composition comprising:

5 [A] a base polymer having a reactivity with the following curing agent [B],

[B] a curing agent which has a group having a reactivity with a hydroxyl group and which has a reactivity with the base polymer [A], and

[C] a polyorthoester prepared by reacting:

10 (a) an orthoester represented by the following Formula (1):



15 wherein R<sup>1</sup> represents a hydrogen atom or an alkyl group having 1 to 4 carbon atoms, and three R<sup>2</sup>'s may be the same or different and each represent an alkyl group having 1 to 4 carbon atoms,

(b) at least one glycol compound selected from α-glycols and β-glycols, and

20 (c) a hydroxyl group-containing compound having at least two hydroxyl groups in a molecule other than the above (b).

[Claim 2] The curable composition as set forth in Claim 1, wherein the base polymer [A] is a hydroxyl group-containing polymer having a hydroxyl group value falling in a range of 20 to 300 mg KOH/g and a number average molecular weight falling in a range of 1,000 to 30,000.

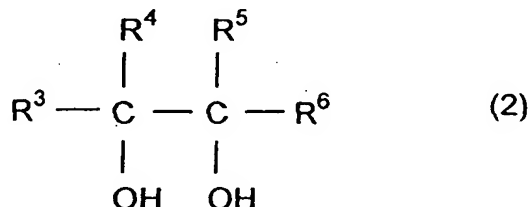
25 [Claim 3] The curable composition as set forth in Claim 1 or 2, wherein the base polymer [A] is at least one polymer selected from acrylic resins, polyester resins and epoxy resins.

[Claim 4] The curable composition as set forth in any one of Claims 1 to 3, wherein the curing agent [B] is at least one compound selected from

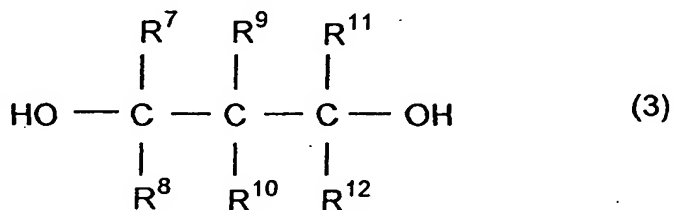
polyisocyanate compounds, amino resins, epoxy group-containing compounds, alkoxysilyl group-containing compounds and compounds having two or more carboxylic anhydride groups.

[Claim 5] The curable composition as set forth in any one of Claims 1 to 4, wherein the orthoester (a) is at least one compound selected from methyl orthoformate, ethyl orthoformate, methyl orthoacetate and ethyl orthoacetate.

[Claim 6] The curable composition as set forth in any one of Claims 1 to 5, wherein the glycol compound (b) is at least one glycol compound selected from an  $\alpha$ -glycol represented by the following Formula (2):



wherein  $\text{R}^3$ ,  $\text{R}^4$ ,  $\text{R}^5$  and  $\text{R}^6$  may be the same or different and each represent a hydrogen atom, an alkyl group having 1 to 24 carbon atoms, an aralkyl group having 7 to 24 carbon atoms or a phenyl group, or a group obtained by substituting a part of these groups with an oxygen atom, and the total of the carbon atoms in the groups represented by  $\text{R}^3$ ,  $\text{R}^4$ ,  $\text{R}^5$  and  $\text{R}^6$  falls in a range of 0 to 24; and  $\text{R}^4$  and  $\text{R}^5$  may form a cyclic structure together with carbon atoms to which they are bonded directly, and a  $\beta$ -glycol represented by the following Formula (3):



wherein  $\text{R}^7$ ,  $\text{R}^8$ ,  $\text{R}^9$ ,  $\text{R}^{10}$ ,  $\text{R}^{11}$  and  $\text{R}^{12}$  may be the same or different and each represent a hydrogen atom, an alkyl group having 1 to 24 carbon atoms, an

aralkyl group having 7 to 24 carbon atoms or a phenyl group, or a group obtained by substituting a part of these groups with an oxygen atom, and the total of the carbon atoms in the groups represented by  $R^7$ ,  $R^8$ ,  $R^9$ ,  $R^{10}$ ,  $R^{11}$  and  $R^{12}$  falls in a range of 0 to 24; and  $R^7$  and  $R^9$  or  $R^7$ ,  $R^9$  and  $R^{11}$  may form a

5 cyclic structure together with carbon atoms to which they are bonded directly.

[Claim 7] The curable composition as set forth in any one of Claims 1 to 6, wherein the glycol compound (b) is at least one compound selected from ethylene glycol, 1,2-propylene glycol, 1,2-hexanediol, neopentyl glycol, 2-methyl-1,3-propanediol, 2-methyl-2,4-pentanediol, 3-methyl-1,3-butanediol,  
10 2-ethyl-1,3-hexanediol, 2,2-diethyl-1,3-propanediol, 2,2,4-trimethyl-1,3-pentanediol and 2-butyl-2-ethyl-1,3-propanediol.

[Claim 8] The curable composition as set forth in any one of Claims 1 to 7, wherein the hydroxyl group-containing compound (c) has a molecular weight falling in a range of 90 to 50,000 and a hydroxyl group value falling in a range of  
15 100 to 1,850.

[Claim 9] The curable composition as set forth in any one of Claims 1 to 8, wherein a hydroxyl group contained in the hydroxyl group-containing compound (c) is blocked by an orthoester of a 5-membered ring or a 6-membered ring constituted by the orthoester (a) and the glycol compound (b).

20 [Claim 10] The curable composition as set forth in any one of Claims 1 to 9, comprising the base polymer [A], the curing agent [B] and the polyorthoester [C] in the following range: the component [A] is 20 to 89 parts by weight, the component [B] is 5 to 70 parts by weight and the component [C] is 1 to 40 parts by weight, based on 100 parts by weight of the total of solid matters contained  
25 in the respective components [A], [B] and [C].

[Claim 11] The curable composition as set forth in any one of Claims 1 to 10, further comprising an acid catalyst.

[Detailed Description of the Invention]

[0001]

30 [Technical field to which the invention belongs]

The present invention relates to a novel curable composition containing a base polymer, a curing agent and a polyorthoester, more specifically to a curable composition which can be reduced in viscosity and

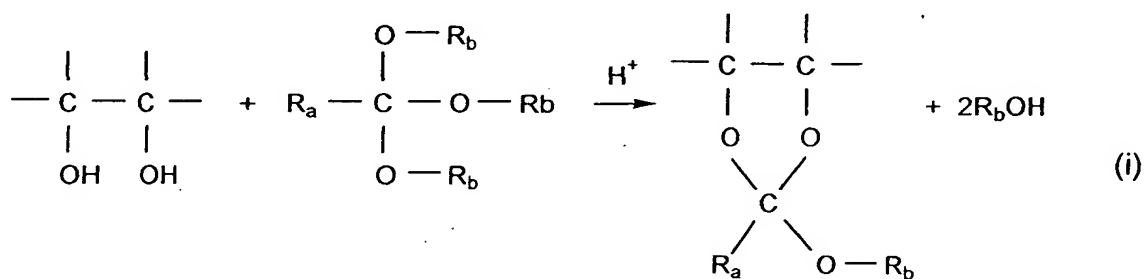
increased in solid content.

[0002]

[Prior art and its problem]

- 5 An orthoester has so far been used as a dehydrating agent and a synthetic raw material for various compounds. Further, an orthoester is known as a protective group for a hydroxyl group and protects a hydroxyl group on such moderate conditions as room temperature in the presence of an acid catalyst by reaction, for example, as shown in the following Equation (i):

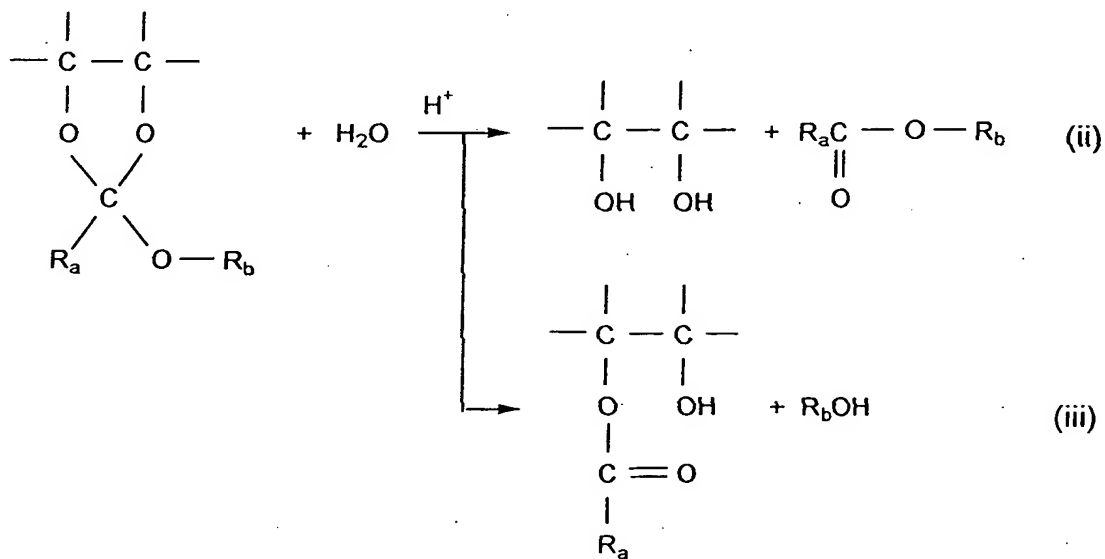
[0003]



10

The protected hydroxyl group is stable under a base condition, but the protective group is readily desorbed under an acid condition by hydrolysis as shown in the following Equation (ii) or (iii):

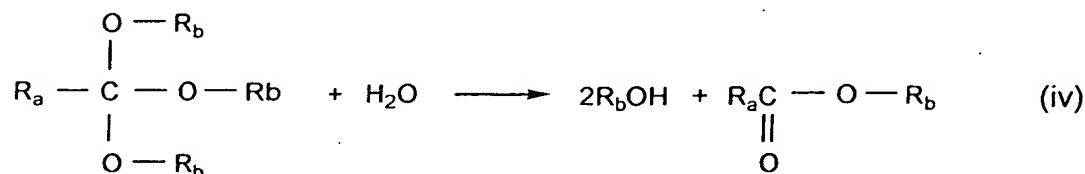
15 [0004]





In general, an orthoester is easily hydrolyzed to form two molecules of alcohol and one molecule of ester as shown in the following Equation (iv):

[0005]



5

Proposed are several techniques on a polyorthoester industrially making use of such characteristics of the polyorthoester described above. For example, a polyorthoester for a photoresist is described in Japanese Patent Publication No.20325/1988, and a polyorthoester for drug delivery is described in Japanese Patent Application Laid-Open No.502465/1993.

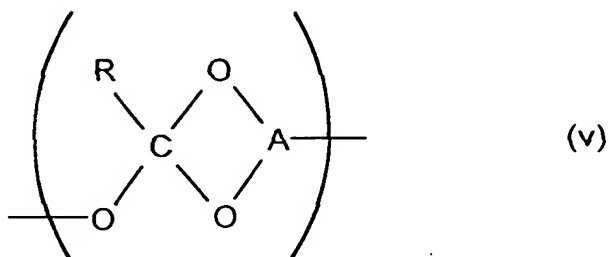
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[0006]

The polyorthoesters described in these official gazettes are compounds which are obtained by condensing triols with orthoesters and which have a specific repetitive unit, for example, a repetitive unit represented by the following Formula (v). However, in producing this compound, only specific triol can be used as a hydroxy group-containing compound, and it has a degree of a freedom only in such an extent that a molecular weight can be changed by a blending ratio of the triol to the orthoester, so that there is the problem that a degree of freedom in molecular design is low.

15

20 [0007]



Further, a polymer having a spiroorthoester structure is described in Japanese Patent Application Laid-Open No.42724/1982, and it is shown that the above polymer is a cross-linking high polymer having a small volumetric

25

shrinkage. However, caprolactone is essentially used as a raw material, and therefore, a degree of freedom in molecular design is low. Furthermore, a polymer having a bicycloorthoester structure is described in Japanese Patent Application Laid-Open No.233114/1985, and it is shown that the above polymer is a cross-linking high polymer having an excellent balance between an elastic modulus and a toughness. However, trimethylolpropane or trimethylolethane is essentially used as a raw material, and therefore, involved therein is the problem that a degree of freedom in molecular design is low.

[0008]

It is known, as described above, that an alkoxy group of an orthoester is subjected to alcohol exchange reaction with a hydroxyl group in the presence of an acid catalyst, and a 5-membered ring, a 6-membered ring or a bicyclo ring can be formed by using a hydroxyl group-containing compound in which two hydroxyl groups are close. Making use of this property, an orthoester is used as a protective group for close hydroxyl groups mainly in the biochemical field.

[0009]

Three alkoxy groups of an orthoester can be subjected to alcohol exchange reaction, and it can be turned into a polyorthoester by combining with polyhydric alcohol. In this case, if an orthoester is subjected merely to exchange reaction with polyhydric alcohol (condensation reaction by dealcohol), it is gelatinized by converting into three dimension.

[0010]

Further, a curable resin composition prepared by combining a polyhydric hydroxyl group-containing compound or a resin with a curing agent has so far been known, and the above curable resin composition has the problems that it is increased in viscosity which originates in a hydroxyl group, reduced in compatibility, and increased in polarity and deteriorated in wetting property when coated on a base material.

[0011]

An object of the present invention is to provide a curable composition using a polyorthoester having a high degree of freedom in molecular design and a low viscosity and capable of being readily controlled in a molecular weight, in

which there involved are no problems on a curing property of the curable composition and a reduction in the hardness; the viscosity can be reduced, and the solid content can be elevated; and the compatibility and the wetting property to a base material are good.

5 [0012]

[Means to solve the problems]

The present inventors repeated intensive investigations in order to solve the above problems. As a result, they found that the above problems could be solved by combining a base polymer; a polyorthoester prepared by  
10 reacting a specific glycol compound, an orthoester and a polyhydric hydroxyl group-containing compound; and a curing agent, and they have come to complete the present invention.

[0013]

That is, the present invention provides a curable composition  
15 comprising:

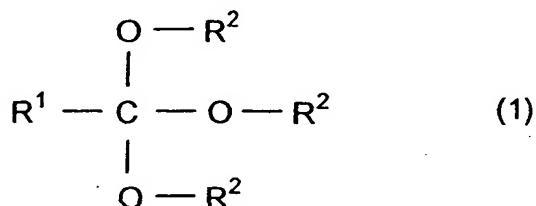
[A] a base polymer having a reactivity with the following curing agent [B],

[B] a curing agent which has a group having a reactivity with a hydroxyl group and which has a reactivity with the base polymer [A], and

20 [C] a polyorthoester prepared by reacting:

(a) an orthoester represented by the following Formula (1):

[0014]



25 wherein  $\text{R}^1$  represents a hydrogen atom or an alkyl group having 1 to 4 carbon atoms, and three  $\text{R}^2$ 's may be the same or different and each represent an alkyl group having 1 to 4 carbon atoms,

(b) at least one glycol compound selected from  $\alpha$ -glycols and  $\beta$ -glycols, and

(c) a hydroxyl group-containing compound having at least two hydroxyl groups in a molecule other than the above (b).

[0015]

The curable composition of the present invention shall be explained  
5 below in further details.

[0016]

[Mode for carrying out the invention]

Base polymer [A]:

The base polymer which is the component [A] in the composition of  
10 the present invention is a polymer having a reactivity with the curing agent [B].  
The base polymer [A] shall not specifically be restricted in a kind thereof as long  
as it has a reactive group (for example, a hydroxyl group, an epoxy group, a  
carboxyl group, an alkoxysilyl group, an acid anhydride group and an  
isocyanato group) capable of being reacted with the curing agent [B] and can be  
15 reacted with the curing agent [B] and cured.

[0017]

The representative examples of the base polymer [A] include, for  
example, hydroxyl group-containing polymers having a hydroxyl group value  
falling in a range of 5 to 500 mg KOH/g, preferably 20 to 300 mg KOH/g and a  
20 number average molecular weight falling in a range of 500 to 50,000, preferably  
1,000 to 30,000. Capable of being given as the resin kind of the base polymer  
[A] are, for example, acrylic resins, polyester resins, silicon polyester resins,  
silicon acrylic resins, fluororesins and epoxy resins. Among them, particularly  
the acrylic resins and the polyester resins can suitably be used.

25 [0018]

Capable of being given as the acrylic resin which can be used as the  
base polymer [A] are, for example, the (co)polymers of the following  
polymerizable unsaturated monomers having a reactive group. Preferred are  
the polymers comprising 30% by weight or more of a C<sub>1</sub> to C<sub>24</sub> alkyl  
30 (meth)acrylate unit based on the whole monomer components.

[0019]

The polymerizable unsaturated monomers constituting the acrylic  
resin include, for example, linear, branched or cyclic C<sub>1</sub> to C<sub>24</sub> alkyl

(meth)acrylates such as methyl (meth)acrylate, ethyl (meth)acrylate, n-propyl (meth)acrylate, isopropyl (meth)acrylate, n-butyl (meth)acrylate, isobutyl (meth)acrylate, tert-butyl (meth)acrylate, 2-ethylhexyl acrylate, n-octyl (meth)acrylate, lauryl (meth)acrylate, tridecyl (meth)acrylate, stearyl

5 (meth)acrylate, isobornyl (meth)acrylate, tricyclodecanyl (meth)acrylate and cyclohexyl acrylate; monoesters of polyhydric alcohols and acrylic acid or methacrylic acid such as 2-hydroxyethyl (meth)acrylate, hydroxypropyl (meth)acrylate, 2,3-dihydroxybutyl (meth)acrylate, 4-hydroxybutyl (meth)acrylate and polyethylene glycol mono(meth)acrylate; compounds

10 obtained by subjecting  $\epsilon$ -caprolactone to ring opening reaction with the monoesters described above of the polyhydric alcohols and acrylic acid or methacrylic acid; carboxyl group-containing polymerizable unsaturated monomers such as acrylic acid, methacrylic acid, itaconic acid, maleic acid and crotonic acid; acid anhydride group-containing polymerizable unsaturated

15 monomers such as maleic anhydride and itaconic anhydride; epoxy group-containing polymerizable unsaturated monomers such as glycidyl (meth)acrylate, allyl glycidyl ether and 3,4-epoxycyclohexylmethyl (meth)acrylate; nitrogen-containing (meth)acrylates such as

20 N,N-dimethylaminoethyl (meth)acrylate, N,N-diethylaminoethyl (meth)acrylate, N,N-dimethylaminopropyl (meth)acrylate and N,N-diethylaminopropyl (meth)acrylate; acrylamide and methacrylamide; (meth)acrylamide derivatives such as N,N-dimethylaminoethyl(meth)acrylamide, N,N-diethylaminoethyl(meth)acrylamide, N,N-dimethylaminopropyl(meth)acrylamide,

25 N,N-diethylaminopropyl(meth)acrylamide, N-methylolacrylamide, N-methoxymethylacrylamide and N-methoxybutylacrylamide; isocyanate group-containing polymerizable unsaturated monomers such as isocyanatethyl (meth)acrylate and m-propenyl- $\alpha,\alpha$ -dimethylbenzylisocyanate; alkoxysilyl group-containing polymerizable unsaturated compounds such as

30 vinyltrimethoxysilane, vinyltriethoxysilane, vinylmethyldimethoxysilane, vinylmethyldiethoxysilane,  $\gamma$ -(meth)acryloyloxypropyltrimethoxysilane,  $\gamma$ -(meth)acryloyloxypropylmethyldimethoxysilane,  $\gamma$ -(meth)acryloyloxypropylmethyldiethoxysilane,

$\gamma$ -(meth)acryloyloxypropyltriethoxysilane,  
 $\beta$ -(meth)acryloyloxyethyltrimethoxysilane and  
 $\gamma$ -(meth)acryloyloxybutylphenyldimethoxysilane; vinyl aromatic compounds such  
 as styrene, vinyltoluene and  $\alpha$ -methylstyrene; acrylonitrile, methacrylonitrile,  
 5 vinyl acetate and Beova Monomer (manufactured by Shell Chemical Co., Ltd.).  
 These polymerizable unsaturated monomers can be used alone or in  
 combination of two or more kinds thereof.

[0020]

The polyester resin capable of being used as the base polymer [A]  
 10 includes, for example, oil-free polyester resins and oil-modified alkyd resins;  
 and modified products of these resins, for example, urethane-modified polyester  
 resins, urethane-modified alkyd resins and epoxy-modified polyester resins, and  
 the polyester resins having a hydroxyl group are suited.

[0021]

15 The oil-free polyester resin described above comprises an ester of a  
 polybasic acid component and a polyhydric alcohol component. Mainly used  
 as the polybasic acid component are, for example, at least one dibasic acid  
 selected from phthalic anhydride, isophthalic acid, terephthalic acid,  
 tetrahydrophthalic anhydride, hexahydrophthalic anhydride,  
 20 hexahydroterephthalic anhydride, succinic acid, fumaric acid, adipic acid,  
 sebacic acid and maleic anhydride, and lower alkyl esters of these acids.  
 Capable of being used in combination are, if necessary, monobasic acids such  
 as benzoic acid, crotonic acid and p-t-butylbenzoic acid, and trivalent or higher  
 polybasic acids such as trimellitic anhydride, methylcyclohexenetricarboxylic  
 25 acid and pyromellitic anhydride. Further, mainly used as the polyhydric alcohol  
 component are, for example, dihydric alcohols such as ethylene glycol,  
 diethylene glycol, propylene glycol, 1,4-butanediol, neopentyl glycol,  
 3-methylpentanediol, 1,4-hexanediol, 1,6-hexanediol, 2-butyl-2-ethylpropylene  
 glycol and 1,4-dimethylolcyclohexane. Further, capable of being used in  
 30 combination are, if necessary, trihydric or higher alcohols such as glycerin,  
 trimethylolethane, trimethylolpropane and pentaerythritol. These polyhydric  
 alcohols can be used alone or in a mixture of two or more kinds thereof.  
 Esterification or transesterification between these both components can be

carried out by a conventionally known method. Isophthalic acid, terephthalic acid and lower alkyl esters of these acids are particularly preferred as the acid component.

[0022]

5           The alkyd resins are obtained by reacting the acid components and the alcohol components of the oil-free polyester resins described above with oil fatty acids by a conventionally known method. Capable of being given as the oil fatty acids are, for example, coconut oil fatty acid, soybean oil fatty acid, linseed oil fatty acid, safflower oil fatty acid, tall oil fatty acid, dehydrated castor  
10 oil fatty acid and tung oil fatty acid. An oil length of the alkyd resins is preferably 30% or less, particularly 5 to 20%.

[0023]

          The urethane-modified polyester resins include resins obtained by reacting the oil-free polyester resins described above or oil-free polyester resins  
15 of a low molecular weight obtained by reacting the acid components and the alcohol components used in producing the oil-free polyester resins described above with polyisocyanate compounds by a conventionally known method. Also, the urethane-modified alkyd resin includes resins obtained by reacting the alkyd resins described above or alkyd resins of a low molecular weight obtained  
20 by reacting the respective components used in producing the alkyd resins described above with polyisocyanate compounds by a conventionally known method.

[0024]

          The polyisocyanate compounds which can be used in producing the  
25 urethane-modified polyester resins and the urethane-modified alkyd resins each described above include, for example, hexamethylenediisocyanate, isophoronediiisocyanate, xylenediisocyanate, tolylenediisocyanate, 4,4'-diphenylmethane-diisocyanate, 4,4'-methylenebis(cyclohexylisocyanate) and 2,4,6-triisocyanatotoluene.

30 [0025]

          In general, capable of being suitably used as the urethane-modified resins described above are the resins having such a modification degree that these polyisocyanate compounds constituting the urethane-modified resins

account for 30% by weight or less, particularly 1 to 20% by weight based on the urethane-modified resins.

[0026]

5       The epoxy-modified polyester resins include, for example, reaction products obtained by using polyester resins produced from the respective components used for producing the polyester resins described above and reacting carboxyl groups in the resins with epoxy group-containing resins and reaction products obtained by bonding hydroxyl groups contained in polyester resins with hydroxyl groups contained in epoxy resins via polyisocyanate  
10    compounds by addition, condensation or graft reaction of the polyester resins with the epoxy resins. In general, the epoxy-modified polyester resins have suitably such a modification degree that the epoxy resins account for 0.1 to 30% by weight, particularly 1 to 20% by weight based on the epoxy-modified polyester resins.

15    [0027]

      The silicon acrylic resins which can be used as the base polymer [A] include, for example; resins obtained by subjecting hydroxyl group-containing acrylic resins to condensation reaction with silicon resins having a hydroxyl group or an alkoxyl group by dehydration or dealcohol by heating, if necessary,  
20    in the presence of a reaction catalyst such as a metal alkoxide compound.

[0028]

      The silicon polyester resins which can be used as the base polymer [A] include, for example, resins obtained by subjecting hydroxyl group-containing polyester resins to condensation reaction with silicon resins  
25    having a hydroxyl group or an alkoxyl group by dehydration or dealcohol by heating, if necessary, in the presence of a reaction catalyst such as a metal alkoxide compound.

[0029]

      The silicon resin having a hydroxyl group or an alkoxyl group used  
30    for producing the silicon acrylic resins and the silicon polyester resins each described above can be obtained in the form of commercial products, and capable of being given as the examples thereof are, for example, SH-6018, DC3074, DC3037 and SR2402 (all products described above are manufactured



by Toray Dow Corning Co., Ltd.); KR9218 and X-40-9220 (all products described above are manufactured by Shin-etsu Chemical Co., Ltd.); and TSR165 and XR-31B1763 (all products described above are manufactured by Toshiba Silicon Co., Ltd.).

5 [0030]

In general, a use proportion of the silicon resins described above in the silicon acrylic resins and the silicon polyester resins falls suitably in a range of 1 to 100 parts by weight, particularly 5 to 60 parts by weight per 100 parts by weight of the resin solid matters of the acrylic resins and the polyester resins.

10 [0031]

The fluororesins which can be used as the base polymer [A] include preferably hydroxyl group-containing fluororesins, and capable of being given are, for example, copolymers of fluoroolefins such as vinyl fluoride and hydroxyl group-containing polymerizable unsaturated monomers such as hydroxyethyl vinyl ether with other polymerizable unsaturated monomers such as ethyl vinyl ether.

[0032]

The epoxy resins which can be used as the base polymer [A] include, for example, glycidyl ether type epoxy resins, glycidyl ester type epoxy resins and other glycidyl type epoxy resins; and modified epoxy resins obtained by modifying these epoxy resins with modifying agents such as alkylphenols and fatty acids. These various epoxy resins may be increased in molecular weight with a linking agent in order to obtain a suitable molecular weight.

[0033]

25 The glycidyl ether type epoxy resins described above are epoxy resins having a glycidyl ether group which can be obtained by reacting polyhydric alcohols or polyhydric phenols with epihalohydrins or alkylene oxides. The examples of the polyhydric alcohols described above include ethylene glycol, diethylene glycol, triethylene glycol, polyethylene glycol, propylene glycol, polypropylene glycol, neopentyl glycol, butylene glycol, 1,6-hexanediol, 30 1,4-hexanediol, glycerin, trimethylolethane, trimethylolpropane, pentaerythritol, diglycerin and sorbitol. The examples of the polyhydric phenols described above include 2,2-bis(4-hydroxyphenyl) propane [bisphenol A],

2,2-bis(2-hydroxyphenyl)propane,  
 2-(2-hydroxyphenyl)-2-(4-hydroxyphenyl)propane, halogenated bisphenol A,  
 bis(4-hydroxyphenyl)methane [bisphenol F], 4,4'-dihydroxybenzophenone,  
 tris(4-hydroxyphenyl)propane, 1,1-bis(4-hydroxyphenyl)ethane,  
 5 4,4'-dihydroxybiphenol, resorcin, hydroquinone, tetrahydroxyphenylethane,  
 1,2,3-tris(2,3-epoxypropoxy)propane, novolak type polyhydric phenols and  
 cresol type polyhydric phenols.

[0034]

The glycidyl ester type epoxy resins described above include resins  
 10 obtained by highly polymerizing diglycidyl phthalate, diglycidyl  
 hexahydrophthalate, diglycidyl tetrahydrophthalate and dimeric acid diglycidyl  
 ester by using linking agents.

[0035]

The other glycidyl type epoxy resins described above include, for  
 15 example, resins obtained by highly polymerizing  
 tetraglycidylaminodiphenylmethane and triglycidylisocyanurate by using linking  
 agents.

[0036]

The modified epoxy resins are epoxy resins obtained by modifying  
 20 epoxy resins before modified such as the glycidyl ether type epoxy resins and  
 the glycidyl ester type epoxy resins each described above with modifying  
 agents such as alkylphenols and fatty acids.

[0037]

The linking agents used in order to optimize the molecular weights of  
 25 the various epoxy resins such as the glycidyl ether type epoxy resins, the  
 glycidyl ester type epoxy resins and the other glycidyl type epoxy resins each  
 described above include, for example, polyhydric phenols, polyhydric alcohols,  
 polybasic acids, primary or secondary amines and polyisocyanates. The  
 polyhydric alcohols and polyhydric phenols which have been given above as the  
 30 examples of the raw materials for the glycidyl ether type epoxy resins can be  
 given as the polyhydric phenols and polyhydric alcohols. The polybasic acids  
 include, for example, adipic acid, azelaic acid, sebacic acid, hexahydrophthalic  
 anhydride, tetrahydrophthalic anhydride, isophthalic acid and dimeric acid.

The primary or secondary amines include, for example, ethylamine, n-propylamine, isopropylamine, n-butylamine, monoethanolamine and hexamethylenediamine. The polyisocyanates include, for example,

5 hexamethylenediisocyanate.

[0038]

Curing agent [B]:

The curing agent which is the component [B] in the composition of the present invention is a substance which has a group having a reactivity with  
10 a hydroxyl group and which can be reacted with a hydroxyl group produced by hydrolysis of an orthoester group in the polyorthoester [C] and also reacted with the base polymer [A] to form a cured matter.

[0039]

Capable of being given as the representative example of the curing  
15 agent [B] are, for example, polyisocyanate compounds, amino resins, epoxy group-containing compounds, alkoxysilyl group-containing compounds and compounds having two or more carboxylic anhydride groups.

[0040]

The polyisocyanate compounds described above include all of  
20 compounds in which isocyanato groups (NCO group) are not blocked (hereinafter abbreviated as "non-blocked polyisocyanate compound") and compounds in which isocyanato groups are blocked (hereinafter abbreviated as "blocked polyisocyanate compound").

[0041]

25 The non-blocked polyisocyanate compounds include, for example, aliphatic diisocyanates such as lysine diisocyanate, hexamethylene diisocyanate and trimethylhexane diisocyanate; alicyclic diisocyanates such as hydrogenated xylilene diisocyanate, isophorone diisocyanate, methylcyclohexane-2,4 or 2,6-diisocyanate,  
30 4,4'-methylenebis(cyclohexylisocyanate) and 1,3-(isocyanatomethyl)cyclohexane; aromatic diisocyanates such as tolylene diisocyanate, xylylene diisocyanate and diphenylmethane diisocyanate; organic polyisocyanates themselves including trivalent or more polyisocyanates such as

lysine triisocyanate, adducts of these organic polyisocyanates to polyhydric alcohols, low molecular weight polyester resins or water, cyclic polymers (for example, isocyanurate) of the respective organic diisocyanates themselves described above and biuret type adducts thereof; and copolymers of isocyanato

5 group-containing ethylenically unsaturated compounds such as isocyanatoethyl (meth)acrylate and m-isopropenyl- $\alpha,\alpha$ -dimethylbenzylisocyanate with other ethylenically unsaturated compounds.

[0042]

The blocked polyisocyanate compounds are obtained by blocking the

10 isocyanato groups of the non-blocked polyisocyanate compounds described above with a blocking agent. In this case, capable of being suitably used as the blocking agent are, for example, blocking agents including phenol bases such as phenol, cresol and xylene; lactam bases such as  $\epsilon$ -caprolactam,  $\delta$ -valerolactam,  $\gamma$ -butyrolactam and  $\beta$ -propiolactam; alcohol bases such as

15 methanol, ethanol, n- or i-propyl alcohol, n-, i- or t-butyl alcohol, ethylene glycol monomethyl ether, ethylene glycol monoethyl ether, ethylene glycol monobutyl ether, diethylene glycol monomethyl ether, diethylene glycol monoethyl ether, propylene glycol monomethyl ether and benzyl alcohol; oxime bases such as formamidoxime, acetaldoxime, acetoxime, methyl ethyl ketoxime,

20 diacetylmonoxime, benzophenoneoxime and cyclohexaneoxime; and active methylene bases such as dimethyl malonate, diethyl malonate, ethyl acetoacetate, methyl acetoacetate and acetylacetone. The isocyanato groups of the polyisocyanates can readily be blocked by mixing the non-blocked polyisocyanate compounds with the blocking agents.

25 [0043]

These polyisocyanate compounds can be used alone or in combination of two or more kinds thereof.

[0044]

The amino resins which can be used as the curing agent [B] include,

30 for example, methylol amino resins obtained by reacting aldehydes with amino components such as melamine, urea, benzoguanamine, acetoguanamine, steroguanamine, spiroguanamine and dicyanediarnide. The aldehydes described above include, for example, formaldehyde, paraformaldehyde,

acetaldehyde and benzaldehyde. Further, compounds obtained by etherifying these methanol amino resins with at least one alcohol are included in the amino resins described above. The alcohols used for the etherification include, for example, monohydric alcohols such as methyl alcohol, ethyl alcohol, n-propyl alcohol, isopropyl alcohol, n-butyl alcohol, isobutyl alcohol, 2-ethylbutanol and 2-ethylhexanol. Among them, particularly suited are the melamine resins obtained by etherifying at least a part of the methanol groups of the methanol melamine resins with the monohydric alcohols having 1 to 4 carbon atoms.

[0045]

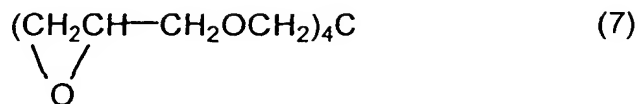
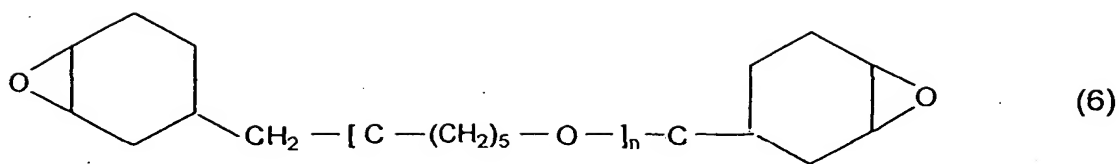
Capable of being given as the specific examples of the melamine resins described above are, for example, methyl-etherified melamine resins such as Cymel 300, ditto 303, ditto 325, ditto 327, ditto 350, ditto 730, ditto 736 and ditto 738 (all described above are manufactured by Mitsui Cytec Co., Ltd.), Melan 522 and ditto 523 (all described above are manufactured by Hitachi Chemical Co., Ltd.), Nikalac MS001, ditto MX430 and ditto MX650 (all described above are manufactured by Sanwa Chemical Co., Ltd.), Sumimal M-55, ditto M-100 and ditto M-40S (all described above are manufactured by Sumitomo Chemical Ind. Co., Ltd.) and Resimine 740 and ditto 747 (all described above are manufactured by Monsanto Co., Ltd.); butyl-etherified melamine resins such as U-van 20SE and ditto 225 (all described above are manufactured by Mitsui Toatsu Co., Ltd.) and Super Beckamine J820-60, ditto L-117-60, ditto L-109-65, ditto 47-508-60, ditto L-118-60 and ditto G821-60 (all described above are manufactured by Dainippon Ink & Chemicals Ind. Co., Ltd.); and methyl ether and butyl ether-mixed etherified melamine resins such as Cymel 232, ditto 266, ditto XV-514 and ditto 1130 (all described above are manufactured by Mitsui Cytec Co., Ltd.), Nikalac MX500, ditto MX600, ditto MS35 and ditto MS95 (all described above are manufactured by Sanwa Chemical Co., Ltd.), Resimine 753 and ditto 755 (all described above are manufactured by Monsanto Co., Ltd.) and Sumimal M-66B (manufactured by Sumitomo Chemical Ind. Co., Ltd.). These melamine resins can be used alone or in combination of two or more kinds thereof.

[0046]

The epoxy group-containing compound which can be used as the

curing agent [B] is a compound having two or more epoxy groups in a molecule, and the representative examples thereof include ethylene glycol diglycidyl ether, polyethylene glycol diglycidyl ether, propylene glycol diglycidyl ether, tripropylene glycol diglycidyl ether, polypropylene glycol diglycidyl ether, 1,4-butanediol diglycidyl ether, neopentyl glycol diglycidyl ether, 1,6-hexanediol diglycidyl ether, glycerin diglycidyl ether, diglycerin tetraglycidyl ether, trimethylolpropane triglycidyl ether, 2,6-diglycidyl phenyl ether, sorbitol triglycidyl ether, triglycidyl isocyanurate, diglycidylamine, diglycidylbenzylamine, diglycidyl phthalate, bisphenol A diglycidyl ether, butadiene dioxide, dicyclopentadiene dioxide, diester of 3,4-epoxycyclohexenecarboxylic acid and ethylene glycol, 3,4-epoxycyclohexylmethyl-3,4-epoxycyclohexanecarboxylate, 3,4-epoxy-6-methylcyclohexylmethyl-3,4-epoxy-6-methylcyclohexanecarboxylate, bis(3,4-epoxycyclohexylmethyl) adipate, glycidyl ether of dicyclopentadieneol epoxide, dipentene dioxide, adducts of bisphenol A type epoxy resins and ethylene oxide, Epoleade GT300 (trifunctional alicyclic epoxy compound, manufactured by Daicel Chemical Industries Ltd.), Epoleade GT400 (tetrafunctional alicyclic epoxy compound, manufactured by Daicel Chemical Industries Ltd.); Epoleade GT301, ditto GT302 and ditto GT303 (ring-opened  $\epsilon$ -caprolactone chain-containing trifunctional alicyclic epoxy compounds, all described above are manufactured by Daicel Chemical Industries Ltd.); Epoleade GT401, ditto GT402 and ditto GT403 (ring-opened  $\epsilon$ -caprolactone chain-containing tetrafunctional alicyclic epoxy compounds, all described above are manufactured by Daicel Chemical Industries Ltd.); Epikote 828, ditto 834 and ditto 1001 (bisphenol A type epoxy resins, all described above are manufactured by Yuka Shell Epoxy Co., Ltd.); Epikote 154 (cresol novolak type epoxy resin, manufactured by Yuka Shell Epoxy Co., Ltd.); Celloxide 2081, ditto 2082 and ditto 2083 each represented by the following Formula (6) (all described above are manufactured by Daicel Chemical Industries Ltd.; in the following Formula [6], the product in which n is 1 is Celloxide 2081, the product in which n is 2 is Celloxide 2082, and the product in which n is 3 is Celloxide 2083); and Denacol EX-411 represented by the following Formula (7) (manufactured by Nagase Kasei Co., Ltd.):

[0047]



5

In Formula (6), n represents an integer of 1 to 3.

[0048]

The epoxy group-containing compound described above includes copolymers of epoxy group-containing polymerizable unsaturated monomers such as glycidyl (meth)acrylate, allyl glycidyl ether and 3,4-epoxycyclohexylmethyl (meth)acrylate with other polymerizable unsaturated monomers. The other polymerizable unsaturated monomers described above include, for example, C<sub>1</sub> to C<sub>24</sub> alkyl (meth)acrylates such as methyl (meth)acrylate, ethyl (meth)acrylate, n-propyl (meth)acrylate, isopropyl (meth)acrylate, n-butyl (meth)acrylate, isobutyl (meth)acrylate, tert-butyl (meth)acrylate, 2-ethylhexyl acrylate, n-octyl (meth)acrylate, lauryl (meth)acrylate, tridecyl (meth)acrylate and stearyl (meth)acrylate; monoesters of polyhydric alcohols and acrylic acid or methacrylic acid such as 2-hydroxyethyl (meth)acrylate, hydroxypropyl (meth)acrylate, 2,3-dihydroxybutyl (meth)acrylate, 4-hydroxybutyl (meth)acrylate and polyethylene glycol mono(meth)acrylate; compounds obtained by subjecting ε-caprolactone to ring opening reaction with the monoesters of the polyhydric alcohols and acrylic acid or methacrylic acid described above; alkoxysilyl group-containing polymerizable unsaturated compounds such as vinyltrimethoxysilane, vinyltriethoxysilane, vinylmethyldimethoxysilane, vinylmethyldiethoxysilane, γ-(meth)acryloyloxypropyltrimethoxysilane, γ-(meth)acryloyloxypropylmethyldimethoxysilane, γ-(meth)acryloyloxypropylmethyldiethoxysilane,

$\gamma$ -(meth)acryloyloxypropyltriethoxysilane,  
 $\beta$ -(meth)acryloyloxyethyltrimethoxysilane and  
 $\gamma$ -(meth)acryloyloxybutylphenyldimethoxysilane; vinyl aromatic compounds such  
 as styrene, vinyltoluene and  $\alpha$ -methylstyrene; acrylonitrile, methacrylonitrile,  
 5 tricyclodecanyl (meth)acrylate, isobornyl (meth)acrylate, vinyl acetate and  
 Beova Monomer (manufactured by Shell Chemical Co., Ltd.).

[0049]

The epoxy group-containing compounds described above can be  
 used alone or in combination of two or more kinds thereof. A content of the  
 10 epoxy group of the epoxy group-containing compound described above shall  
 not specifically be restricted, and the epoxy equivalent falls suitably in a range  
 of usually 100 to 3,000, preferably 100 to 1,500.

[0050]

The alkoxysilyl group-containing compound which can be used as  
 15 the curing agent [B] is a compound having two or more alkoxysilyl groups in a  
 molecule and includes, for example, alkoxysilanes having no polymerizable  
 unsaturated groups such as dimethoxydimethylsilane, dimethoxydiethylsilane,  
 dimethoxydiphenylsilane, diethoxydimethylsilane, trimethoxymethylsilane,  
 trimethoxyethylsilane, trimethoxypropylsilane, trimethoxyphenylsilane,  
 20 tetramethoxysilane, tetraethoxysilane, tetrabutoxysilane and  
 dimethoxydiethoxysilane; polymerizable unsaturated group-containing  
 alkoxysilanes such as vinyltrimethoxysilane, vinyltriethoxysilane,  
 vinylmethyldimethoxysilane, vinylmethyldiethoxysilane,  
 $\gamma$ -(meth)acryloyloxypropyltrimethoxysilane,  
 25  $\gamma$ -(meth)acryloyloxypropylmethyldimethoxysilane,  
 $\gamma$ -(meth)acryloyloxypropylmethyldiethoxysilane,  
 $\gamma$ -(meth)acryloyloxypropyltriethoxysilane,  
 $\beta$ -(meth)acryloyloxyethyltrimethoxysilane and  
 $\gamma$ -(meth)acryloyloxybutylphenyldimethoxysilane; partially condensed products of  
 30 the above alkoxysilanes having no polymerizable unsaturated groups and/or the  
 polymerizable unsaturated group-containing alkoxysilanes described above;  
 and copolymers of the polymerizable unsaturated group-containing  
 alkoxysilanes described above with polymerizable unsaturated monomers



which can be copolymerized with the above alkoxysilanes.

[0051]

The polymerizable unsaturated monomers capable of being copolymerized with the alkoxysilanes which are the monomer components for the copolymers described above include monomers other than the alkoxysilyl group-containing polymerizable unsaturated compounds capable of being used as the "other polymerizable unsaturated monomers" which are copolymerized with the epoxy group-containing polymerizable unsaturated monomers when the epoxy group-containing compounds described above are copolymers.

10 [0052]

Capable of being given as the compound (hereinafter, abbreviated as a "polyacid anhydride") having two or more carboxylic anhydride groups which can be used as the curing agent [B] are, for example, pyromellitic anhydride, a condensation product (ethylenebis[anhydrotrimellitate]) of one mole of ethylene glycol and 2 moles of trimellitic anhydride, and a condensation product (glycerintris[anhydrotrimellitate]) of one mole of glycerin and 3 moles of trimellitic anhydride; linear or cyclic polyacid anhydrides obtained by subjecting polybasic acids such as succinic acid, adipic acid, azelaic acid, sebacic acid, dodecanedioic acid, dimer acid, ethyl-octadecanedioic acid, phenyl-hexadecanedioic acid and 1,4-cyclohexanedicarboxylic acid to intermolecular condensation; and polymers comprising a polymerizable unsaturated acid anhydride such as maleic anhydride and tetrahydrophthalic anhydride as a single monomer component. The monomers other than the polymerizable unsaturated acid anhydrides which can form the above polymers include, for example, alkyl (meth)acrylates having 1 to 24 carbon atoms such as methyl (meth)acrylate, ethyl (meth)acrylate, n-propyl (meth)acrylate, isopropyl (meth)acrylate, n-butyl (meth)acrylate, isobutyl (meth)acrylate, tert-butyl (meth)acrylate, 2-ethylhexyl acrylate, n-octyl (meth)acrylate, lauryl (meth)acrylate, tridecyl (meth)acrylate and stearyl (meth)acrylate; polymerizable unsaturated monocarboxylic acids such as acrylic acid, methacrylic acid and crotonic acid; vinyl aromatic compounds such as styrene, vinyltoluene and  $\alpha$ -methylstyrene; and acrylonitrile, methacrylonitrile, tricyclodecanyl (meth)acrylate, isobornyl (meth)acrylate, vinyl acetate, and Beova Monomer

(manufactured by Shell Chemical Co., Ltd.)

[0053]

A content of the acid anhydride groups in the polyacid anhydrides described above shall not specifically be restricted, and the whole acid value  
 5 based on the acid anhydride groups falls suitably in a range of usually 50 to 1,100 mg KOH/g, preferably 80 to 800 mg KOH/g.

[0054]

The respective curing agents [B] described above can be used alone or in combination of two or more kinds thereof.

10 [0055]

Polyorthoester [C]

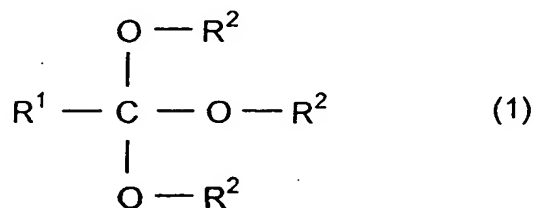
The polyorthoester which is the component [C] in the composition of the present invention is a reaction product of the orthoester (a), the glycol compound (b) and the hydroxyl group-containing compound (c) each described  
 15 below.

[0056]

Orthoester (a):

The orthoester which is the component (a) is a compound represented by the following Formula (1):

20 [0057]



wherein R<sup>1</sup> represents a hydrogen atom or an alkyl group having 1 to 4 carbon atoms, and three R<sup>2</sup>'s may be the same or different and each represent an alkyl  
 25 group having 1 to 4 carbon atoms.

[0058]

In Formula (1) described above, the alkyl group having 1 to 4 carbon atoms represented by R<sup>1</sup> or R<sup>2</sup> is linear or branched and includes, for example, methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl and t-butyl.

[0059]

The specific examples of the orthoester (a) include, for example, methyl orthoformate, ethyl orthoformate, propyl orthoformate, butyl orthoformate, methyl orthoacetate, ethyl orthoacetate, methyl orthopropionate, ethyl orthopropionate, methyl orthobutyrate and ethyl orthobutyrate. Among them, methyl orthoformate, ethyl orthoformate, methyl orthoacetate and ethyl orthoacetate are suited. They may be used alone or in combination of two or more kinds thereof.

[0060]

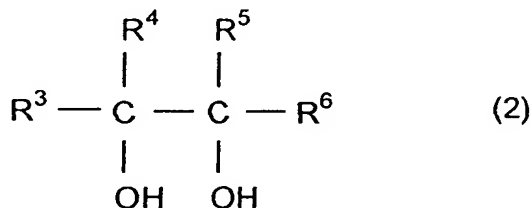
10 Glycol compound (b):

The glycol compound which is the component (b) is at least one glycol compound selected from  $\alpha$ -glycols and  $\beta$ -glycols having two hydroxyl groups in a molecule.

[0061]

15 Among them, a compound represented by the following Formula (2) can suitably be used as the  $\alpha$ -glycol:

[0062]



20 wherein  $R^3$ ,  $R^4$ ,  $R^5$  and  $R^6$  may be the same or different and each represent a hydrogen atom, an alkyl group having 1 to 24 carbon atoms, an aralkyl group having 7 to 24 carbon atoms or a phenyl group, or a group obtained by substituting a part of these groups with an oxygen atom, and the total of the carbon atoms in the groups represented by  $R^3$ ,  $R^4$ ,  $R^5$  and  $R^6$  falls in a range of  
 25 0 to 24, preferably 0 to 10; and  $R^4$  and  $R^5$  may form a cyclic structure together with carbon atoms to which they are bonded directly.

[0063]

In Formula (2) described above, the alkyl group having 1 to 24 carbon atoms represented by  $R^3$ ,  $R^4$ ,  $R^5$  or  $R^6$  is linear, branched or cyclic and

includes methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, t-butyl, pentyl, hexyl, n-octyl, 2-ethylhexyl, decyl, dodecyl, octadecyl, cyclohexyl, methylcyclohexyl, cyclohexylmethyl and cyclohexylethyl.

[0064]

- 5                    In Formula (2) described above, the aralkyl group having 7 to 24 carbon atoms represented by  $R^3$ ,  $R^4$ ,  $R^5$  or  $R^6$  is preferably an alkyl group substituted with phenyl, and the specific examples thereof include benzyl and phenethyl.

[0065]

- 10                   In Formula (2) described above, the group obtained by substituting a part of the alkyl group, the aralkyl group or the phenyl group, represented by  $R^3$ ,  $R^4$ ,  $R^5$  or  $R^6$ , with an oxygen atom includes, for example, an alkoxyalkyl group such as methoxymethyl, ethoxymethyl, propoxymethyl and butoxymethyl; an alkanoyloxyalkyl group such as acetoxymethyl and acetoxylethyl; and an  
15                   aryloxyalkyl group such as phenoxymethyl and phenoxyethyl.

[0066]

Among them,  $R^3$ ,  $R^4$ ,  $R^5$  or  $R^6$  in Formula (2) described above is preferably a hydrogen atom or an alkyl group having 1 to 6 carbon atoms.

[0067]

- 20                   In Formula (2) described above, the cyclic structure which can be formed by  $R^4$  and  $R^5$  together with carbon atoms to which they are bonded directly includes, for example, a cyclohexyl group and a cyclopentyl group.

[0068]

- 25                   Thus, the representative examples of the  $\alpha$ -glycols include, for example, ethylene glycol, 1,2-propylene glycol, 1,2-butylene glycol, 2,3-butylene glycol, 1,2-hexanediol, 1,2-dihydroxycyclohexane, pinacol and hydrolysis products of long chain alkyl monoepoxides; fatty acid monoglycerides ( $\alpha$  products) such as glycerin monoacetate ( $\alpha$  product) and glycerin monostearate ( $\alpha$  product); and 3-ethoxypropane-1,2-diol and 3-phenoxypropane-1,2-diol.

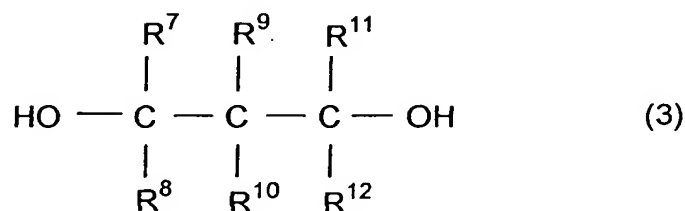
- 30                   Among them, ethylene glycol, 1,2-propylene glycol and 1,2-hexanediol are suited.

[0069]

On the other hand, particularly a compound represented by the

following Formula (3) can suitably be used as the  $\beta$ -glycol:

[0070]



- 5 wherein  $R^7$ ,  $R^8$ ,  $R^9$ ,  $R^{10}$ ,  $R^{11}$  and  $R^{12}$  may be the same or different and each represent a hydrogen atom, an alkyl group having 1 to 24 carbon atoms, an aralkyl group having 7 to 24 carbon atoms or a phenyl group, or a group obtained by substituting a part of these groups with an oxygen atom, and the total of the carbon atoms in the groups represented by  $R^7$ ,  $R^8$ ,  $R^9$ ,  $R^{10}$ ,  $R^{11}$  and
- 10  $R^{12}$  falls in a range of 0 to 24; and  $R^7$  and  $R^9$  or  $R^7$ ,  $R^9$  and  $R^{11}$  may form a cyclic structure together with carbon atoms to which they are bonded directly.

[0071]

- In Formula (3) described above, the alkyl group having 1 to 24 carbon atoms represented by  $R^7$ ,  $R^8$ ,  $R^9$ ,  $R^{10}$ ,  $R^{11}$  or  $R^{12}$  includes the same ones as those described above as the alkyl group represented by  $R^3$ ,  $R^4$ ,  $R^5$  or  $R^6$  in
- 15 Formula (2) described above.

[0072]

- In Formula (3) described above, the aralkyl group having 7 to 24 carbon atoms represented by  $R^7$ ,  $R^8$ ,  $R^9$ ,  $R^{10}$ ,  $R^{11}$  or  $R^{12}$  is preferably an alkyl group substituted with phenyl, and the specific examples thereof include benzyl and phenethyl.
- 20

[0073]

- In Formula (3) described above, the group obtained by substituting a part of the alkyl group, the aralkyl group or the phenyl group, represented by  $R^7$ ,  $R^8$ ,  $R^9$ ,  $R^{10}$ ,  $R^{11}$  or  $R^{12}$ , with an oxygen atom includes, for example, an alkoxyalkyl group such as methoxymethyl, ethoxymethyl, propoxymethyl and butoxymethyl; an alkanoyloxyalkyl group such as acetoxymethyl and acetoxylethyl; and an aryloxyalkyl group such as phenoxymethyl and phenoxyethyl.
- 25

[0074]

In Formula (3) described above, the cyclic structure which can be formed by  $R^7$  and  $R^9$  or  $R^7$ ,  $R^9$  and  $R^{11}$  together with carbon atoms to which they are bonded directly includes, for example, a cyclohexyl group and a  
 5 cyclopentyl group.

[0075]

Among them,  $R^7$ ,  $R^8$ ,  $R^9$ ,  $R^{10}$ ,  $R^{11}$  or  $R^{12}$  in Formula (3) described above is preferably a hydrogen atom or an alkyl group having 1 to 6 carbon atoms.

10 [0076]

Thus, the representative examples of the  $\beta$ -glycols include, for example, neopentyl glycol, 2-methyl-1,3-propanediol, 2-methyl-2,4-pentanediol, 3-methyl-1,3-butanediol, 2-ethyl-1,3-hexanediol, 2,2-diethyl-1,3-propanediol, 2,2,4-trimethyl-1,3-pentanediol, 2-butyl-2-ethyl-1,3-propanediol,  
 15 2-phenoxypropane-1,3-diol, 2-methyl-2-phenylpropane-1,3-diol, 1,3-propylene glycol, 1,3-butylene glycol, dimethylolpropionic acid, dimethylolbutanoic acid, 2-ethyl-1,3-octanediol and 1,3-dihydroxycyclohexane; and fatty acid monoglycerides ( $\beta$  products) such as glycerin monoacetate ( $\beta$  product) and glycerin monostearate ( $\beta$  product). Among them, suited are neopentyl glycol,  
 20 2-methyl-1,3-propanediol, 2-methyl-2,4-pentanediol, 3-methyl-1,3-butanediol, 2-ethyl-1,3-hexanediol, 2,2-diethyl-1,3-propanediol, 2,2,4-trimethyl-1,3-pentanediol and 2-butyl-2-ethyl-1,3-propanediol.

[0077]

(c) Hydroxyl group-containing compound:

25 The hydroxyl group-containing compound which is the component (c) is a compound having at least two hydroxyl groups in a molecule other than the glycol compound (b) described above.

[0078]

The hydroxyl group-containing compound (c) includes compounds  
 30 having two hydroxyl groups other than  $\alpha$ -glycols and  $\beta$ -glycols, or compounds having 3 or more hydroxyl groups in a molecule.

[0079]

The compounds having two hydroxyl groups other than  $\alpha$ -glycols and

$\beta$ -glycols include, for example, 1,4-butanediol, 1,4-dihydroxycyclohexane,  
 1,5-pentanediol, 1,6-hexanediol, 2,5-hexanediol, 3-methyl-1,5-pentanediol,  
 1,4-dimethylolcyclohexane, tricyclodecanedimethanol,  
 2,2-dimethyl-3-hydroxypropyl-2,2-dimethyl-3-hydroxypropionate (this compound  
 5 corresponds to ester of hydroxypivalic acid and neopentyl glycol), bisphenol A,  
 bisphenol F, bis(4-hydroxyhexyl)-2,2-propane, bis(4-hydroxyhexyl)methane,  
 3,9-bis(1,1-dimethyl-2-hydroxyethyl)-2,4,8,10-tetraoxaspiro[5,5]undecane,  
 diethylene glycol, triethylene glycol, tetra- or more polyethylene glycol,  
 dipropylene glycol, tripropylene glycol, tetra- or more polypropylene glycol,  
 10 copolymers having hydroxyl groups at both terminals obtained by  
 copolymerizing ethylene oxide with propylene oxide, linear polyesters having  
 hydroxyl groups at both terminals such as polycaprolactonediol,  
 polycarbonatediol and carboxylic acid adducts of diepoxide.  
 [0080]

15 The compounds having 3 or more hydroxyl groups described above  
 include, for example, glycerin, diglycerin, triglycerin, pentaerythritol,  
 dipentaerythritol, sorbitol, mannitol, trimethylolethane, trimethylolpropane,  
 ditrimethylolpropane, tris(2-hydroxyethyl)isocyanurate, gluconic acid and  
 polymers having 3 or more hydroxyl groups (polyesters, polyethers, acryl  
 20 polymers, ketone resins, phenol resins, epoxy resins, urethane resins those  
 having 3 or more hydroxyl groups; polyvinyl alcohols which are saponified  
 products of polyvinyl acetates; and natural saccharides such as glucose).  
 [0081]

25 Capable of being suitably used as the hydroxyl group-containing  
 compound (c) are the compounds having a hydroxyl group value falling in a  
 range of 20 to 1,850 mg KOH/g, particularly 40 to 1,650 mg KOH/g.

#### Production of polyorthoester [C]:

30 In producing the polyorthoester [C] which is one component of the  
 composition of the present invention, a blending ratio of the orthoester (a), the  
 glycol compound (b) and the hydroxyl group-containing compound (c) shall not  
 specifically be restricted. In general, it is suitable, in terms of an easiness in  
 controlling the molecular weight, to use the orthoester (a) in a proportion falling  
 in a range of 0.01 to 10 moles, preferably 0.05 to 5 moles and more preferably

0.1 to 2 moles; and the glycol compound (b) in a proportion falling in a range of 0.01 to 10 moles, preferably 0.05 to 5 moles and more preferably 0.1 to 2 moles, each per equivalent of a hydroxyl group contained in the hydroxyl group-containing compound (c).

5 [0082]

The polyorthoester [C] can be obtained by subjecting the three components (a), (b) and (c) described above to condensation reaction. For example, it can suitably be produced by heating the three components described above at a temperature falling in a range of usually room temperature  
10 to 250°C, preferably 70 to 200°C for approximately 1 to 20 hours, if necessary, in the presence of an organic solvent and an acid catalyst to subject them to condensation reaction.

[0083]

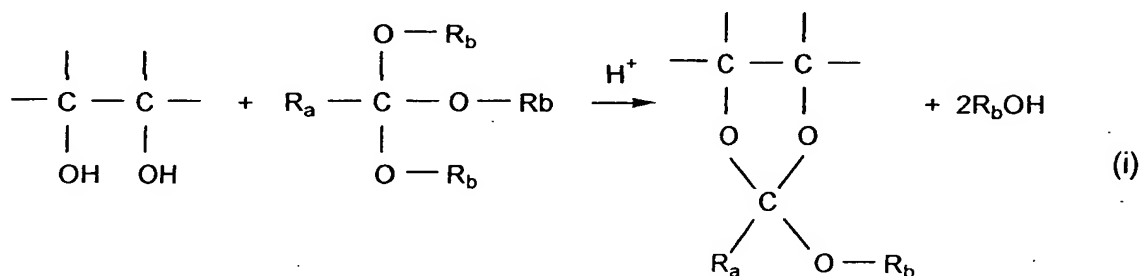
In the reaction described above, an alkoxy group of the orthoester  
15 (a) causes exchange reaction with the alcohol parts of the glycol compound (b) and the hydroxyl group-containing compound (c). In this case, the orthoester (a) is preferentially reacted with the  $\alpha$ -glycol or  $\beta$ -glycol which is the glycol compound (b) having hydroxyl groups close to each other to form a cyclic structure. That is, the glycol compound (b) is cyclized by preferentially reacting  
20 with two functional groups (alkoxyl groups) of the orthoester (a) which is trifunctional. The remaining one alkoxyl group of the orthoester is reacted with the hydroxyl group-containing compound (c). Thus, cross-linking between the molecules is not brought about in producing the polyorthoester [C], and therefore, the product can be inhibited from being increased in molecular weight  
25 and viscosity. On the other hand, if the orthoester (a) is reacted directly with the hydroxyl group-containing compound (c) in the absence of the glycol compound (b), cross-linking between the molecules takes place, so that the product is rapidly increased in molecular weight and viscosity. The polyorthoester [C] in the composition of the present invention is a product which  
30 is inhibited from being increased in molecular weight and viscosity by further adding the glycol compound (b) to the orthoester (a) and the hydroxyl group-containing compound (c) to react them.

[0084]



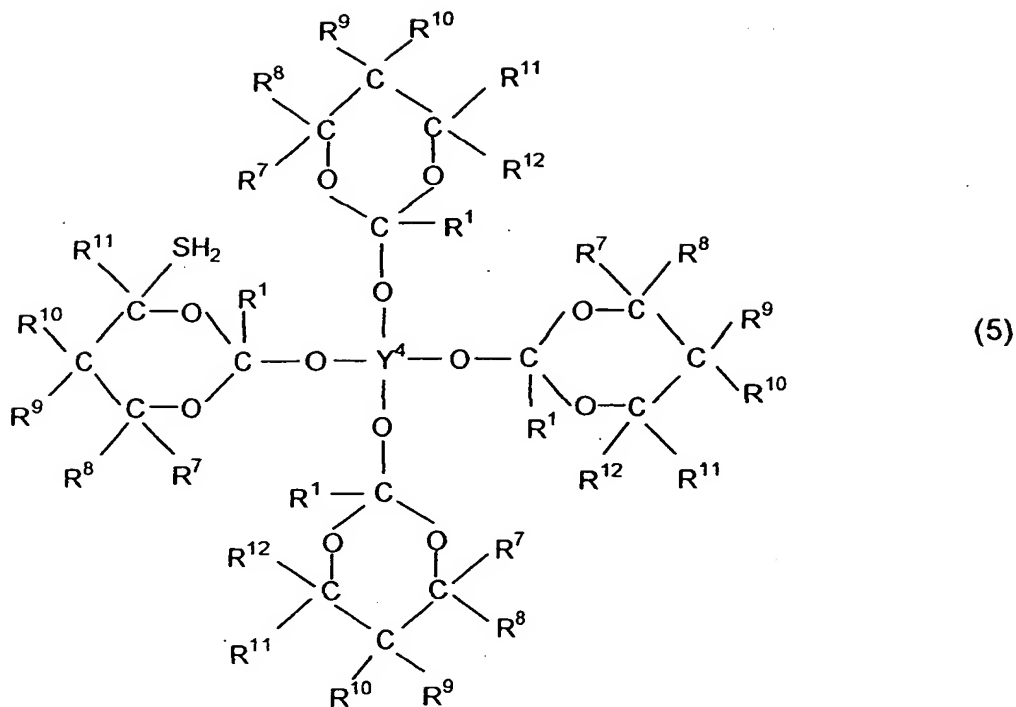
When used as the raw materials are, for example, the orthoester represented by Formula (1) described above, the  $\alpha$ -glycol represented by Formula (2) described above and the compound having 2 hydroxyl groups in a molecule, the polyorthoester [C] produced in the manner described above can have a structure represented by the following Formula (4):

[0085]



wherein  $Y^1$  represents a residue obtained by removing, from the compound having 2 hydroxyl groups in a molecule, the 2 hydroxyl groups; and  $R^1$ ,  $R^3$ ,  $R^4$ ,  $R^5$  and  $R^6$  are the same as defined above. Also, when used as the raw materials are, for example, the orthoester represented by Formula (1) described above, the  $\beta$ -glycol represented by Formula (3) described above and the compound having 4 hydroxyl groups in a molecule, the polyorthoester [C] produced in the manner described above can have a structure represented by the following Formula (5):

[0086]



wherein  $Y^2$  represents a residue obtained by removing, from the compound having four hydroxyl groups in a molecule, the four hydroxyl groups; and  $R^1$ ,  $R^7$ ,  $R^8$ ,  $R^9$ ,  $R^{10}$ ,  $R^{11}$  and  $R^{12}$  are the same as defined above.

[0087]

#### Curable composition of the present invention

The curable composition of the present invention comprises the base polymer [A], the curing agent [B] and the polyorthoester [C], each described above, as the essential components. A blending proportion of the above components [A], [B] and [C] shall not specifically be restricted, and usually, it falls suitably in the following range based on 100 parts by weight of the total solid matters contained in the components [A], [B] and [C]:

[0088]

component [A]: 20 to 89 parts by weight, preferably 25 to 87 parts by weight, component [B]: 5 to 70 parts by weight, preferably 10 to 60 parts by weight, and component [C]: 1 to 40 parts by weight, preferably 3 to 25 parts by weight.

[0089]

The curable composition of the present invention can suitably contain, if necessary, an acid catalyst, an organic solvent, a curing catalyst, a

pigment, a UV absorber, a coated surface-controlling agent, an antioxidant, a fluidity-controlling agent and a wax in addition to the components [A], [B] and [C] described above. The acid catalyst described above is a catalyst for accelerating a reaction of deblocking an orthoester group to reproduce a hydroxyl group. The kind thereof shall not specifically be restricted, and capable of being used as well are, for example, inorganic acids such as hydrochloric acid, sulfuric acid and nitric acid; sulfonic acid compounds such as methanesulfonic acid, ethanesulfonic acid, paratoluenesulfonic acid, dodecylbenzenesulfonic acid, dinonylnaphthalenesulfonic acid and dinonylnaphthalenedisulfonic acid; compounds obtained by neutralizing the sulfonic acid compounds described above with bases such as amines; esters of the sulfonic acid compounds described above with primary, secondary or tertiary alcohols such as n-propanol, n-butanol, n-hexanol, n-octanol, isopropanol, 2-butanol, 2-hexanol, 2-octanol, cyclohexanol and tert-butanol;  $\beta$ -hydroxyalkylsulfonic acid esters obtained by reacting the sulfonic acid compounds described above with oxirane group-containing compounds such as glycidyl acetate and butyl glycidyl ether; carboxylic acids such as formic acid, acetic acid, propionic acid, butyric acid, 2-ethylhexanoic acid and octanoic acid; organic phosphoric acid base compounds such as monobutyl phosphate, dibutyl phosphate, monoisopropyl phosphate, diisopropyl phosphate, monoethyl phosphate, dioctyl phosphate, monodecyl phosphate, didecyl phosphate, metaphosphoric acid, orthophosphoric acid, pyrophosphoric acid, trimethyl phosphate, triethyl phosphate, tributyl phosphate, trioctyl phosphate, tributoxyethyl phosphate, tris-chloroethyl phosphate, triphenyl phosphate and tricresyl phosphate; light latent acid-generating agents which generate acids by irradiating with UV rays such as Cyracure UVI-6970, ditto UVI-6974 and ditto UVI-6990 (all described above are manufactured by Union Carbide Co., Ltd., U.S.A.), Irgacure 261 and ditto 264 (all described above are manufactured by Ciba Specialty Chemicals Co., Ltd.), CIT-1682 (manufactured by Nippon Soda Co., Ltd.), BBI-102 (manufactured by Midori Kagaku Co., Ltd.) and Adeka Optomer SP-150 and ditto SP-170 (all described above are manufactured by Asahi Denka Kogyo K.K.); and Lewis acids.

[0090]

The composition of the present invention can usually be a composition of a non-solvent or organic solvent type. When it is the composition of an organic solvent type, solvents which can dissolve or disperse the respective components of the composition of the present invention can be used, and capable of being given are, for example, hydrocarbon base solvents such as heptane, toluene, xylene, octane and mineral spirits; ester base solvents such as ethyl acetate, n-butyl acetate, isobutyl acetate, ethylene glycol monomethyl ether acetate and diethylene glycol monobutyl ether acetate; ketone base solvents such as methyl ethyl ketone, methyl isobutyl ketone, diisobutyl ketone and cyclohexanone; alcohol base solvents such as methanol, ethanol, isopropanol, n-butanol, sec-butanol and isobutanol; ether base solvents such as n-butyl ether, dioxane, ethylene glycol monomethyl ether and ethylene glycol monoethyl ether; and aromatic petroleum base solvents such as Swasol 310, Swasol 1000 and Swasol 1500 (all described above are manufactured by Cosmo Oil Co., Ltd.) and Shellsol A (manufactured by Shell Chemical Co., Ltd.). These organic solvents can be used alone or in combination of two or more kinds thereof.

[0091]

The curing catalyst described above which is blended, if necessary, with the composition of the present invention is intended for accelerating the curing reaction of the composition, and suited when the curing agent [B] is a blocked polyisocyanate compound is, for example, a curing catalyst which promotes the dissociation of a blocking agent in the blocked polyisocyanate compound which is a curing agent. Capable of being given as the suited curing catalyst are, for example, organic metal catalysts such as tin octanoate, dibutyltin di(2-ethylhexanoate), dioctyltin di(2-ethylhexanoate), dioctyltin diacetate, dibutyltin dilaurate, dibutyltin oxide, dioctyltin oxide and lead 2-ethylhexanoate.

[0092]

When the curing agent [B] is an amino resin such as a melamine resin, particularly a methyl-etherified or methyl ether and butyl ether-mixed etherified melamine resin having a low molecular weight, phosphoric acid, a sulfonic acid compound or an amine-neutralized product of the sulfonic acid

compound is suitably used as the curing catalyst. The representative examples of the sulfonic acid compound include p-toluenesulfonic acid, dodecylbenzenesulfonic acid, dinonylnaphthalenesulfonic acid and dinonylnaphthalenedisulfonic acid. The amine in the amine-neutralized product of the sulfonic acid compound may be any one of primary amines, secondary amines and tertiary amines.

[0093]

The curing catalyst used when the curing agent [B] is the epoxy group-containing compound includes, for example, chelating compounds such as tetrakis(acetylacetonato)zirconium, cobalt acetylacetonate, tris(acetylacetonato)aluminum and manganese acetylacetonate; chelating reaction products of compounds having a  $\beta$ -hydroxyamino structure with lead (II) oxide; metal carboxylates such as lead 2-ethylhexanoate, lead secanoate, lead naphthenoate, lead octanoate, lead acetate, lead lactate and zirconium octanoate; and imidazole compounds such as imidazole, 2-methylimidazole, 2-isopropylimidazole, 2-undecylimidazole and 2-phenylimidazole.

[0094]

The curing catalyst used when the curing agent [B] is the alkoxysilyl group-containing compound includes, for example, organic sulfonic acid compounds such as dodecylbenzenesulfonic acid, paratoluenesulfonic acid, dinonylnaphthalenesulfonic acid and trifluorosulfonic acid; amine-neutralized products of these organic sulfonic acid compounds; and phosphoric acid base compounds such as monobutyl phosphate, dibutyl phosphate, monoisopropyl phosphate, diisopropyl phosphate, monooctyl phosphate, dioctyl phosphate, monodecyl phosphate, didecyl phosphate, metaphosphoric acid, orthophosphoric acid, pyrophosphoric acid, trimethyl phosphate, triethyl phosphate, tributyl phosphate, trioctyl phosphate, tributoxyethyl phosphate, tris-chloroethyl phosphate, triphenyl phosphate and tricresyl phosphate.

[0095]

The curing catalyst used when the curing agent [B] is the polyacid anhydride includes, for example, quaternary salt catalysts such as tetraethylammonium bromide, tetrabutylammonium bromide, tetraethylammonium chloride, tetrabutylammonium fluoride,

tetrabutylphosphonium bromide, triphenylbenzylphosphonium chloride and n-dodecyltributylammonium bromide; and amines such as triethylamine and tributylamine.

[0096]

5           Capable of being given as the pigment compounded, if necessary, with the composition of the present invention are, for example, inorganic color pigments such as titanium white, carbon black, red iron oxide and titanium yellow; organic color pigments such as quinacridone red, azo red, phthalocyanine blue, phthalocyanine green and organic yellow pigments; brilliant pigments such as aluminum powder and brilliant mica powder; extender pigments such as silica powder, calcium carbonate, barium sulfate, mica, clay and talc; and rust preventive pigments such as calcium ion-exchanged silica, phosphate base rust preventive pigments and chromate base pigments.

[0097]

15           The curing conditions of the curable composition of the present invention can be changed according to the kind of the curing agent [B] and shall not specifically be restricted. Usually, when the curing time is 5 minutes or longer, it is suitable to be room temperature (0°C) to 200°C, particularly 60°C to 180°C, and when the curing time is shorter than 5 minutes, it is preferable to be 20 60°C to 300°C, particularly 80°C to 260°C.

[0098]

The composition of the present invention can suitably be used for uses in coating material compositions, adhesives and inks, particularly as a coating material composition.

25 [0099]

[Examples]

The present invention shall more specifically be explained below with reference to examples and comparative examples, and "parts" and "%" mean "parts by weight" and "% by weight" respectively.

30 [0100]

#### Synthesis of acrylic resin solution

##### Synthesis Example 1

A reactor equipped with a stirrer, a condenser, a

temperature-controlling device, a nitrogen-introducing tube and a dropping funnel was charged with 983 parts of xylene and 240 parts of 3-methoxybutyl acetate, and the reactor was substituted with nitrogen, heated and maintained at 135°C. Added drop by drop thereto in 4 hours was a mixture comprising

5 600 parts of styrene, 384 parts of isobutyl methacrylate, 480 parts of 2-ethylhexyl acrylate, 552 parts of 2-hydroxyethyl methacrylate, 450 parts of Placel FM-3X (80% xylene solution, manufactured by Daicel Chemical Industries, Ltd.), 24 parts of acrylic acid and 120 parts of azobisisobutyronitrile. After finishing dropping, the solution was ripened at 135°C for 30 minutes, and

10 then, a mixed solution comprising 144 parts of xylene and 24 parts of azobisisobutyronitrile was added drop by drop in one hour. Then, the solution was maintained at 135°C for 30 minutes and diluted with 144 parts of xylene to obtain an acrylic resin solution (A-1) having a non-volatile matter content of about 60% and a Gardner viscosity (20°C) of T. The resulting resin (solid

15 matter) had a hydroxyl value of about 117 mg KOH/g, a number average molecular weight of 3,400 and a weight average molecular weight of 7,500.

[0101]

#### Synthesis Example 2

A reactor equipped with a stirrer, a condenser, a

20 temperature-controlling device, a nitrogen-introducing tube and a dropping funnel was charged with 983 parts of xylene and 240 parts of 3-methoxybutyl acetate, and the reactor was substituted with nitrogen, heated and maintained at 135°C. Added drop by drop thereto in 4 hours was a mixture comprising 240 parts of styrene, 240 parts of methyl methacrylate, 720 parts of n-butyl

25 methacrylate, 240 parts of 2-hydroxyethyl methacrylate, 960 parts of glycidyl methacrylate and 120 parts of azobisisobutyronitrile. After finishing dropping, the solution was ripened at 135°C for 30 minutes, and then, a mixed solution comprising 144 parts of xylene and 24 parts of azobisisobutyronitrile was added drop by drop in one hour. Then, the solution was maintained at 135°C for 30

30 minutes and diluted with 234 parts of xylene to obtain an acrylic resin solution (A-2) having a non-volatile matter content of about 60% and a Gardner viscosity (20°C) of RS. The resulting resin (solid matter) had a hydroxyl value of about 43 mg KOH/g, an epoxy equivalent of about 360, a number average molecular

weight of about 3,000 and a weight average molecular weight of 6,900.

[0102]

### Synthesis Example 3

A reactor equipped with a stirrer, a condenser, a  
5 temperature-controlling device, a nitrogen-introducing tube and a dropping  
funnel was charged with 288 parts of xylene and 432 parts of 3-methoxybutyl  
acetate, and the reactor was substituted with nitrogen, heated and maintained  
at 135°C. Added drop by drop thereto in 4 hours was a mixture comprising  
701 parts of styrene, 463 parts of isobutyl methacrylate, 636 parts of isobutyl  
10 acrylate, 600 parts of maleic anhydride and 720 parts of 3-methoxybutyl acetate,  
and a mixture comprising 264 parts of t-butylperoxy-2-ethylhexanoate and 672  
parts of xylene was added drop by drop at the same time therewith in 4 hours  
and 30 minutes to obtain an acrylic resin solution (A-3) having a non-volatile  
matter content of about 56% and a Gardner viscosity (20°C) of ST. The  
15 resulting resin (solid matter) had a whole acid value of about 270, a number  
average molecular weight of about 1,900 and a weight average molecular  
weight of about 4,100.

[0103]

### Synthesis of polyester solution

#### 20 Synthesis Example 4

A reactor equipped with a stirrer, a condenser, a  
temperature-controlling device, a water separator, a fractionating column, a  
nitrogen-introducing tube and a solvent-recovering device was charged with 200  
parts of neopentyl glycol, 101 parts of trimethylolpropane, 132 parts of  
25 isophthalic acid, 116 parts of adipic acid, 61 parts of hexahydrophthalic  
anhydride and 68 parts of 1,4-cyclohexanedicarboxylic acid, and the reactor  
was substituted with nitrogen and then started to be heated. The temperature  
was elevated at a fixed rate from 170°C to 230°C in 3 hours while removing  
condensation water and then maintained at 230°C for one hour. Thereafter, 30  
30 parts of xylene was added thereto, and the reaction was further promoted for 7  
hours while maintaining at 230°C and removing condensation water by means  
of the water separator. Then, the reactor was cooled down, and 370 parts of  
xylene was added thereto to obtain a polyester solution (E-1) having a



non-volatile matter content of about 60% and a Gardner viscosity (20°C) of V. The resulting resin (solid matter) had a resin acid value of about 10 mg KOH/g, a hydroxyl value of about 128 mg KOH/g, a number average molecular weight of 2,300 and a weight average molecular weight of 12,000.

5 [0104]

Production of polyorthoester

Production Example 1

A reactor equipped with a stirrer, a condenser, a temperature-controlling device and a solvent-recovering device was charged  
10 with 424 parts of methyl orthoformate, 640 parts of 2-butyl-2-ethyl-1,3-propanediol, 136 parts of pentaerythritol and 4 parts of a 90% formic acid aqueous solution and maintained at about 85°C for one hour while distilling off methanol produced by alcohol exchange reaction. Then, the temperature was elevated up to 190°C in 2 hours, and 365 parts of methanol  
15 was recovered to obtain a colorless and liquid polyorthoester (C-1). The polyorthoester (C-1) thus obtained had a Gardner viscosity of X<sup>+</sup> and a weight average molecular weight of about 1,540.

[0105]

Production Example 2

20 A reactor equipped with a stirrer, a condenser, a temperature-controlling device and a solvent-recovering device was charged with 480 parts of methyl orthoacetate, 640 parts of 2-butyl-2-ethyl-1,3-propanediol, 136 parts of pentaerythritol and 4 parts of a 90% formic acid aqueous solution and maintained at about 85°C for one hour  
25 while distilling off methanol produced by alcohol exchange reaction. Then, the temperature was elevated up to 190°C in 2 hours, and 360 parts of methanol was recovered to obtain a colorless and liquid polyorthoester (C-2). The polyorthoester (C-2) thus obtained had a Gardner viscosity of M and a weight average molecular weight of about 1,060.

30 [0106]

Production Example 3

A reactor equipped with a stirrer, a condenser, a temperature-controlling device and a solvent-recovering device was charged

with 480 parts of methyl orthoacetate, 416 parts of neopentyl glycol, 136 parts of pentaerythritol and 4 parts of a 90% formic acid aqueous solution and maintained at about 85°C for one hour while distilling off methanol produced by alcohol exchange reaction. Then, the temperature was elevated up to 190°C in 2 hours, and 366 parts of methanol was recovered to obtain a colorless and liquid polyorthoester (C-3). The polyorthoester (C-3) thus obtained had a Gardner viscosity of G<sup>+</sup> and a weight average molecular weight of about 430. [0107]

#### Production Example 4

A reactor equipped with a stirrer, a condenser, a temperature-controlling device and a solvent-recovering device was charged with 360 parts of methyl orthoacetate, 312 parts of neopentyl glycol, 550 parts of Placel 305 (polycaprolactonepolyol, manufactured by Daicel Chemical Industries, Ltd.) and 3 parts of a 90% formic acid aqueous solution and maintained at about 85°C for one hour while distilling off methanol produced by alcohol exchange reaction. Then, the temperature was elevated up to 190°C in 2 hours, and 267 parts of methanol was recovered to obtain a colorless and liquid polyorthoester (C-4). The polyorthoester (C-4) thus obtained had a Gardner viscosity of S and a weight average molecular weight of about 1,640. [0108]

#### Production Example 5

A reactor equipped with a stirrer, a condenser, a temperature-controlling device and a solvent-recovering device was charged with 636 parts of methyl orthoformate, 960 parts of 2-butyl-2-ethyl-1,3-propanediol, 254 parts of dipentaerythritol and 2 parts of a 90% formic acid aqueous solution and maintained at about 85°C for one hour while distilling off methanol produced by alcohol exchange reaction. Then, the temperature was elevated up to 190°C in 2 hours, and 553 parts of methanol was recovered to obtain a colorless and liquid polyorthoester (C-5). The polyorthoester (C-5) thus obtained had a Gardner viscosity of Z<sub>4</sub> and a weight average molecular weight of about 2,010. [0109]

The polyorthoesters thus obtained in Production Examples 1 to 5

were all colorless and liquid polyorthoesters and had a solid content of substantially about 100%.

[0110]

Preparation of curable composition

5 Example 1

The 60% acrylic resin solution (A-1) 16.7 parts obtained in Synthesis Example 1 was blended with 9.9 parts of Desmodur N-3300 (triisocyanurate of hexamethylenediisocyanate, manufactured by Sumitomo Bayer Urethane Co., Ltd.), 3.0 parts of the polyorthoester (C-1) obtained in Production Example 1, 10 4.0 parts of xylene, 0.7 part of 3-methoxybutyl acetate and 0.9 part of Nacure 5543 (sulfonic acid base acid catalyst solution, active ingredient: about 25%, manufactured by King Industries Inc., U.S.A.), and the solution was homogeneously mixed to obtain a curable composition having a non-volatile matter content of 65%.

15 [0111]

Examples 2 to 11 and Comparative Examples 1 to 5

The same operation as in Example 1 was carried out to obtain the respective curable compositions, except that in Example 1, the blending composition was changed as shown in the following Table 1. The curable 20 compositions obtained in Example 11 and Comparative Example 5 had a non-volatile matter content of 60%, and all the curable compositions obtained in Examples 2 to 10 and Comparative Examples 1 to 4 had a non-volatile matter content of 65%.

[0112]

25 The remark in Table 1 means the following:

(Remark 1) Cymel 303: methyl-etherified melamine resin, solid content: about 100%, manufactured by Mitsui Cytec Co., Ltd.

[0113]

The respective curable compositions obtained in the above 30 Examples 1 to 11 and Comparative Examples 1 to 5 were tested for a gel ratio, a coating film hardness and a solvent resistance of the cured coating films based on the following test methods. The test results thereof are shown in the following Table 1. Further, the Gardner viscosities of the respective curable

compositions obtained in Examples 1 to 11 and Comparative Examples 1 to 5 are shown as well in Table 1. All the respective curable compositions were excellent in compatibility and wetting property to the base material.

[0114]

## 5 Test methods

Gel ratio:

The curable composition was coated on a teflon sheet so that a dried film thickness was about 30  $\mu\text{m}$  and dried at 140°C for 30 minutes, and the coating film was peeled off to obtain a free coating film. The free coating film  
10 was extracted under refluxing in acetone for 6 hours, and the gel ratio (%) was determined from the coating film weights before and after extraction according to the following equation:

[0115]

$$15 \quad \text{gel ratio (\%)} = \frac{(\text{coating film weight after extraction})}{(\text{coating film weight before extraction})} \times 100$$

Coating film hardness:

20 The curable composition was coated on a polished cold rolled steel plate so that a dried film thickness was about 30  $\mu\text{m}$  and dried at 140°C for 30 minutes to obtain a coating film. The coating film thus obtained was measured for a Knoop hardness at 20°C on the conditions of a load of 25 g and a load-applying time of 20 seconds by means of a microhardness meter ("Model  
25 DMH-2", a product manufactured by Matsuzawa Seiki Co., Ltd.). The larger the value is, the higher the hardness becomes.

[0116]

Solvent resistance:

The curable composition was coated on a polished cold rolled steel  
30 plate so that a dried film thickness was about 30  $\mu\text{m}$  and dried at 140°C for 30 minutes to obtain a coating film. The surface thereof was rubbed in a length of about 5 cm at a load of about 1  $\text{kg}/\text{cm}^2$  by 20 reciprocations with a three-folded gauze impregnated with xylene, and then the state of the coated face was observed and evaluated according to the following criteria:

[0117]

O : coating film has no scratches and dull glossiness and is of a good state

Δ : coating film has slightly scratches or slightly dull glossiness and is of a little inferior state

× : coating film is dissolved or markedly scratched

[0118]

[Table 1]

Table 1

	Example											Comparative Example				
	1	2	3	4	5	6	7	8	9	10	11	1	2	3	4	5
Component [A] Base polymer	Acrylic resin solution A-1	16.7	16.7	16.7	16.7	16.7	16.7					16.7	16.7			
	Acrylic resin solution A-2										16.7					16.7
	Polyester solution E-1							16.7	16.7	16.7				16.7	16.7	
Component [B] Curing agent	Desmodur N-3300	9.9	9.5	8.7	9.2	9.7		10.3				4.1		4.5		
	Cymel 303 (remark 1)					4	4		4	4			4		4	
	Acrylic resin solution A-3										16.7					16.7
Component [C] Polyorthoester	Production Example 1 (C-1)	3				3		3	3		3					
	Production Example 2 (C-2)		3				3									
	Production Example 3 (C-3)			2						3						
	Production Example 4 (C-4)				4											
	Production Example 5 (C-5)					3										

To be continued

[illegible]

Respective comparisons of Example 1 with Comparative Example 1, Example 6 with Comparative Example 2, Example 8 with Comparative Example 3, Example 9 with Comparative Example 4 and Example 11 with Comparative Example 5 show that in all cases, the addition of the polyorthoesters allow the curable compositions to be reduced in viscosity in the same non-volatile matter content and solvent composition without lowering the curing property and the hardness.

[0119]

It can be found from the matters described above that the curable composition according to the present invention can achieve a reduction in viscosity without reducing the coating film performances by adding the polyorthoester and therefore is useful particularly as a binder for a high solid coating material.

[0120]

[Effect of the invention]

The curable composition of the present invention is a composition comprising a base polymer, a curing agent and a polyorthoester. The polyorthoester comprises an orthoester structure introduced into a hydroxyl group part of a hydroxyl group-containing compound and has a high degree of freedom in molecular design, and it is free of problems originating in a hydroxyl group and possible to reduce in viscosity.

[0121]

The curable composition of the present invention comprises a polyorthoester in addition to a system of a base polymer and a curing agent, and therefore, the system can be reduced in viscosity and elevated in solid content; it is free of problems originating in a hydroxyl group, for example, a problem on a rise in viscosity of the curable composition; and it is possible to be increased in solid content and to be excellent in compatibility, wetting property to a base material and storage stability.



[Abstract]

[Subject]

The present invention is to provide a curable composition which is free of problems originating in a hydroxyl group, for example, a problem on a rise in viscosity of the curable composition; possible to be increased in solid content; and excellent in compatibility, wetting property to a base material and storage stability.

[Means for Solution]

A curable composition comprising [A] a base polymer, [B] a curing agent which has a group having a relativity with a hydroxyl group and which has a relativity with the base polymer [A], and [C] a polyorthoester which is prepared by reacting (a) an orthoester such as methyl orthoformate, ethyl orthoformate, methyl orthoacetate or ethyl orthoacetate, (b) at least one glycol compound selected from  $\alpha$ -glycols and  $\beta$ -glycols, and (c) a hydroxyl group-containing compound having at least two hydroxyl groups in a molecule other than the above (b).

[Selected Drawing]

None